

Half-Metallic Ferromagnetism in the Heusler Compound Co_2FeSi revealed by Resistivity, Magnetoresistance, and Anomalous Hall Effect measurements

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We present electrical transport data for single-crystalline Co_2FeSi which provide clear-cut evidence that this Heusler compound is truly a half-metallic ferromagnet, i.e. it possesses perfect spin-polarization. More specifically, the temperature dependence of ρ is governed by electron scattering off magnons which are thermally excited over a sizeable gap $\Delta \approx 100$ K (~ 9 meV) separating the electronic majority states at the Fermi level from the unoccupied minority states. As a consequence, electron-magnon scattering is only relevant at $T \gtrsim \Delta$ but freezes out at lower temperatures, i.e., the spin-polarization of the electrons at the Fermi level remains practically perfect for $T \lesssim \Delta$. The gapped magnon population has a decisive influence on the magnetoresistance and the anomalous Hall effect (AHE): i) The magnetoresistance changes its sign at $T \sim 100$ K, ii) the anomalous Hall coefficient is strongly temperature dependent at $T \gtrsim 100$ K and compatible with Berry phase related and/or side-jump electronic deflection, whereas it is practically temperature-independent at lower temperatures.

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The class of Heusler compounds offers a tool box for studying various materials functionalities and interesting physical ground states [1–3]. For example, for some Co_2 -based Heusler compounds full spin polarization at the Fermi level referred to as half-metallic ferromagnetism [3–6] has been predicted [see fig. 1a for an illustration of the density of states (DOS) of a half-metallic ferromagnet (HMF)], which renders these materials prime candidates for spintronics applications. Most prominent among these materials is the Heusler compound Co_2FeSi , since this ferromagnetic metal possesses the highest Curie temperature ever found in Heusler compounds ($T_C = 1100$ K) together with a large magnetic moment of $m = 6 \mu_B/\text{f.u.}$ [7].

In this letter we present the electronic transport properties, viz., resistivity, magnetoresistance and Hall effect, of high quality Co_2FeSi single crystals [8]. Our data provide fresh evidence of Co_2FeSi being a true HMF: At high temperatures ($T \gtrsim 100$ K), the resistivity ρ is governed by scattering off ferromagnetic magnons. This magnon scattering process is exponentially suppressed at lower temperatures which implies a complete suppression of the minority electron density of states at the Fermi level where the excitation gap for these levels is extracted from resistivity as $\Delta \sim 100$ K. This crucial finding is further corroborated by a sign change of magnetoresistance from negative at high temperatures ($T \gtrsim 110$ K) to positive at lower temperatures. Furthermore, we observe that the activation of ferromagnetic magnons across Δ has a profound impact on the anomalous Hall effect (AHE). While the anomalous Hall coefficient is practically temperature independent at $T \lesssim 100$ K, consistent with dominating extrinsic skew scattering or the ordinary Lorentz-force in-

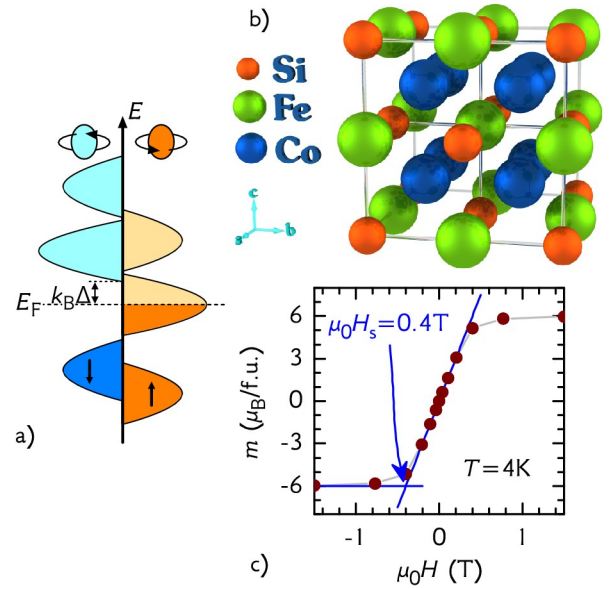


FIG. 1: a) $L2_1$ crystal structure of Co_2FeSi . b) Sketch of the density of states of a HMF in dependence of energy for minority and majority electrons with energy gap at Fermi level E_F for minority electrons. c) Magnetic moment of Co_2FeSi single crystal.

duced Hall effect, it strongly increases at higher T , where it follows $\sim \rho^2$, suggestive of dominating Berry phase and/or side-jump contributions caused by activated ferromagnetic magnons.

Experimental details – Large single crystals of Co_2FeSi , which crystallizes in the $L2_1$ structure with a unit cell size of $a = 5.64$ Å [9] (fig. 1b), were prepared using the floating zone technique [8]. Magnetization measurements

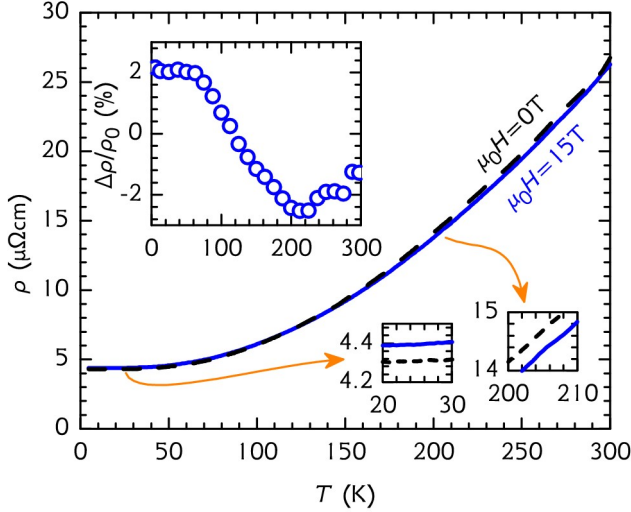


FIG. 2: Temperature dependence of the resistivity in zero and 15 T magnetic field. Upper left inset: Difference ($\Delta\rho = \rho(15\text{T}) - \rho(0\text{T})$, $\rho_0 = \rho(0\text{T})$) of these measurements. Lower right insets: Close-up to low and high temperatures.

confirm the predicted magnetization of $m = 6\mu_B/\text{f.u.}$ and yield a saturation field of $\mu_0 H_s = 0.4\text{ T}$ (see fig. 1c). The susceptibility below H_s is $\chi = 5.4$. Due to the high Curie temperature, the temperature dependences of the saturation field and susceptibility are negligible up to room temperature. For resistivity measurements in magnetic field, a single crystal with dimensions of $a = b = c = 3\text{ mm}$ was used. Resistivity was measured using a standard 4-probe alternating current dc-technique. Hall measurements were done simultaneously using 2 additional Hall voltage contacts.

Resistivity – Fig. 2 shows the resistivity ρ as a function of temperature T for zero magnetic field and for $\mu_0 H = 15\text{ T}$. In zero field the resistivity shows a clear metallic temperature dependence with a monotonic increase with increasing temperature. A residual resistivity of $\rho_R \approx 4\mu\Omega\text{cm}$ and a residual resistivity ratio of $\rho(300\text{K})/\rho_R = 6.5$ shows the extraordinary quality of our samples.

The resistivity of metals is usually dominated by electron-phonon scattering. This typically leads to a linear temperature dependence at elevated temperatures ($T \gtrsim 50\text{ K}$) and a saturation towards residual resistivity at lower temperatures. Such a linear part is completely absent in Co_2FeSi . However, for ferromagnetic materials like the compound under scrutiny, another scattering process may be found, i.e., electron-magnon scattering, which often leads to a quadratic temperature dependence of the resistivity (see e.g. [10–15]).

However, the attempt to fit $\rho(T)$ of Co_2FeSi with a quadratic and a quadratic plus linear temperature dependence (for pure magnon and magnon/phonon scattering of the electrons, respectively) fails completely (see fig. 3). Thus, the conventional electron-magnon scatter-

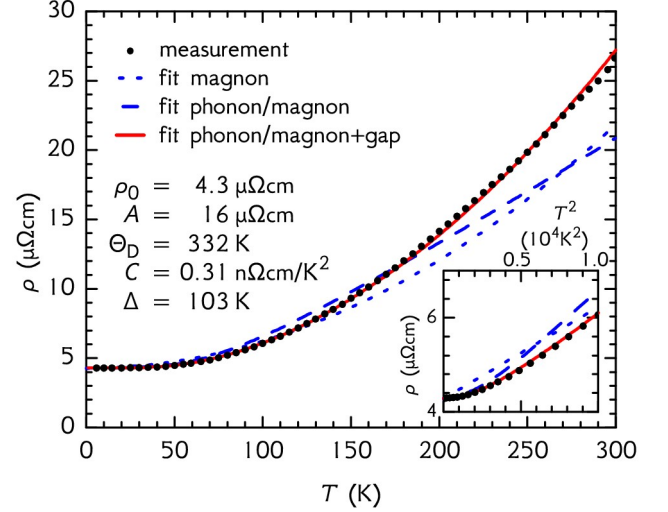


FIG. 3: Resistivity in dependence of the temperature. Measured values are displayed as black dots. Blue chopped lines show fits with magnonic ($\sim T^2$) and magnonic/phononic temperature dependence. The red continuous line shows a fit where an additional Boltzmann factor was added to the magnonic scattering, its fitting parameters are displayed (see text for formulas).

ing of band ferromagnets cannot account for the observed temperature dependence of the resistivity. It can, however, be rationalized much better if the predicted half-metallic character of the material is taken into account. In HMF, the minority spin electronic states are completely gapped at the Fermi level, where $k_B\Delta$ is the minimum excitation energy of majority charge carriers to occupy empty minority states involving a spin flip (see fig. 1b). As a consequence, at low temperatures ($T \lesssim \Delta$) the generation of magnons (which involves a spin flip) and in particular electron-magnon scattering must be exponentially suppressed. At higher temperatures ($T \gtrsim \Delta$) spin flips and magnon scattering would become possible when the Fermi distribution smears out the occupation of energy states around the Fermi level.

In order to test against this scenario we fitted the resistivity with combined scattering of a residual (ρ_R), magnonic (ρ_M) and phononic (ρ_P) contribution:

$$\rho(T) = \rho_R + \rho_M(T) + \rho_P(T). \quad (1)$$

The residual resistivity ρ_R is temperature independent and caused by defects of the ideal crystal lattice. The usually quadratic magnonic term (ρ_M) was weighted by a Boltzmann factor:

$$\rho_M(T) = CT^2 \cdot e^{-\Delta/T}. \quad (2)$$

The parameter C is a measure of the strength of the magnon scattering process. The phononic part of the scattering process (ρ_P) is described by the Bloch-

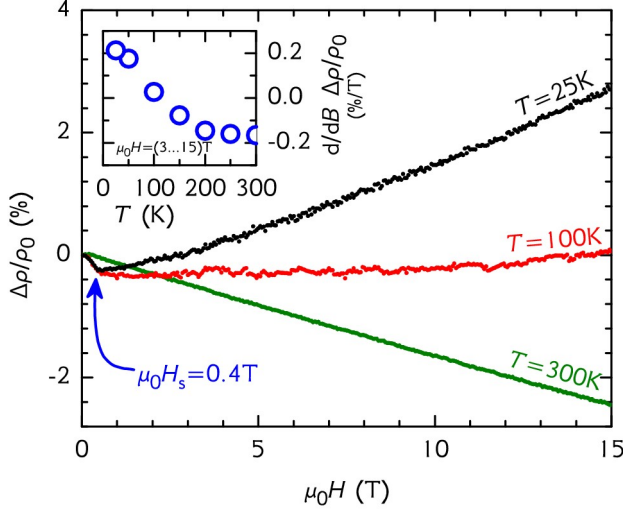


FIG. 4: Magnetic field dependence of resistivity at different temperatures. Inset: slope of $\rho(B)$ in the applied field range of $\mu_0 H = (3 \dots 15)$ T.

Grüneisen formula:

$$\rho_P(T) = A \left(\frac{T}{\Theta_D} \right)^5 \int_0^{\Theta_D/T} \frac{x^5}{(e^x - 1)(1 - e^{-x})} dx. \quad (3)$$

With the Debye temperature $\Theta_D = 332$ K, determined from specific heat measurements, the resistivity fit ¹ yields an energy gap of

$$\Delta = 103 \text{ K}, (k_B \Delta = 8.9 \text{ meV})^2. \quad (4)$$

It is worth to point out that the substantial size of the gap implies that the spin-polarization of the electrons at the Fermi level remains practically perfect for $T \lesssim \Delta$.

Magnetoresistance – An independent verification of the afore derived scenario of gapped-out magnon scattering can be obtained from the corresponding magnetoresistance of Co_2FeSi , because the influence of a magnetic field on the magnons should be reflected by the resistivity. As can be seen in fig. 2, a magnetic field of 15 T yields a relatively small ($\sim 2\%$) but well resolvable change of the resistivity as compared to the zero-field data. The inset highlights the sign and the temperature dependence of $\Delta\rho = \rho(15\text{T}) - \rho(0)$. Apparently $\Delta\rho > 0$ ($\Delta\rho < 0$) for $T \lesssim 110$ K ($T \gtrsim 110$ K).

To get a more detailed view of this dependence on the magnetic field we measured the resistivity while sweeping the magnetic field and maintaining the temperatures (fig. 4). The inset of fig. 4 shows the slope of the resistivity for fields higher than the saturation field. Up to the

saturation field of $\mu_0 H_s = 0.4$ T a small decrease of the resistivity can be seen, which can naturally be explained by alignment and joining of magnetic domains which decreases the probability of domain wall scattering. For high fields $H > H_s$, the resistivity decreases with increasing field at high temperatures; at low temperatures it increases with increasing field.

This temperature dependence of the magnetoresistance can perfectly be explained by the afore presented scenario of the resistivity being dominated by scattering of spin-polarized electrons off gapped magnons. The negative magnetoresistance at high temperatures signals a decrease of the electronic scattering with increasing field. This is consistent with ferromagnetic magnons being the main scattering centers for electrons since a magnetic field enlarges the magnon energy and thus lowers the number of magnons. Upon lowering the temperature, magnons in a HMF are expected to freeze out exponentially since only one spin state is present at the Fermi level. Hence electron-defect scattering must become important at lower temperatures and one expects a more conventional positive magnetoresistance. Indeed, such a behaviour is observed at $T \lesssim 110$ K: $\Delta\rho > 0$ and $\Delta\rho(B)$ approaches a positive quadratic field dependence, which is characteristic for conventional multi-band metals.

Hall effect – Having established Co_2FeSi being a HMF and activated magnon generation for $T \gtrsim 100$ K, we now move on to the anomalous Hall effect (AHE) of Co_2FeSi . The Hall resistivity of this compound is shown in fig. 5 along with its corresponding Hall coefficients in the inset. The Hall resistivity $\rho_{xy}(\mu_0 H)$ is characteristic for the AHE in ferromagnetic materials: it has a linear dependence on the applied magnetic field but with a strong kink and a changing slope just at the saturation field of $\mu_0 H_s = 0.4$ T. Since the Hall effect depends on the magnetic field H as well as on its magnetization M , one considers two Hall coefficients, viz. the ordinary Hall coefficient R and the so-called anomalous Hall coefficient R_A , which connects the applied magnetic field H and the magnetization M of the sample, respectively, with the Hall resistivity [15, 16]:

$$\rho_{xy} = \mu_0 (R \cdot H + R_A \cdot M). \quad (5)$$

Thus, from our Hall data in high magnetic fields the ordinary Hall coefficient R can be obtained, and with the knowledge of this coefficient and $\chi = 5.4$, the anomalous Hall coefficient R_A can be extracted from the low-field data, where $M = \chi \cdot H$. The results are shown in the inset of fig. 5. The ordinary Hall coefficient R is small and does not change much with temperature as expected for a metal. It slightly decreases with increasing temperature which indicates a change of the mobility of the charge carriers consistent with the multi-band nature of this material [7]. At low temperatures the anomalous Hall coefficient R_A is comparable to the ordinary one, while at $T \gtrsim 100$ K it strongly increases with T .

¹ Note that electron-phonon scattering can be left out if one fits the data only up to 200 K.

² We estimate the uncertainty to less than 30%.

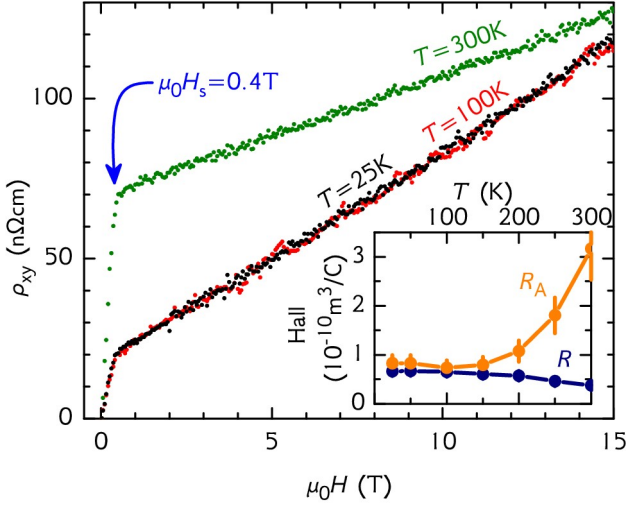


FIG. 5: Hall resistivity in dependence of the applied magnetic field for different temperatures. Inset: ordinary Hall coefficient R and anomalous Hall coefficient R_A in dependence of the temperature.

The huge increase of R_A cannot be explained by Lorentz force deflection, and implies the relevance of fundamental electronic deflection mechanisms known in ferromagnetic materials. More specifically, following the classification given by Nagaosa et al. [16], one expects on the one hand $R_A \sim \rho^2$ for the *intrinsic* Berry phase-related contribution to the AHE as well for the so-called *side-jump* scattering contribution. On the other hand, for the extrinsic *skew scattering*, $R_A \sim \rho$ is expected. In order to test the AHE in Co_2FeSi against one of these scenarios, we plot R_A as a function of the measured $\rho(T)$ (fig. 6). Indeed, for ρ larger than the residual resistivity ρ_R , the data very well follow $R_A \propto (\rho - \rho_R)^2 + \text{const}$, which provides clear evidence that the AHE is dominated by the intrinsic Berry phase and/or the side-jump scattering contributions. Interestingly, this resistivity regime corresponds to temperatures $T \gtrsim 100$ K, i.e., to high temperatures where magnons are significantly populated, which may affect both the Berry phase as well as the side-jump scattering. At lower resistivity values, which correspond to the low-temperature regime where magnons apparently freeze out, the observed $\sim \rho^2$ behavior breaks down and R_A becomes independent of ρ , implying that a qualitatively different deflection mechanism becomes dominant. Indeed the comparable magnitude of the anomalous (R_A) and ordinary (R) Hall coefficients for $T \lesssim 100$ K suggests a contribution of the Lorentz force induced Hall effect (see inset fig. 5), but also skew scattering contributions cannot be excluded. Note, that the temperature dependent change of ρ in this regime is very small, and thus R_A cannot be tested against the expected linear dependence of ρ . Impurity dependent investigations of R_A should shed more light on this issue, but are beyond the scope

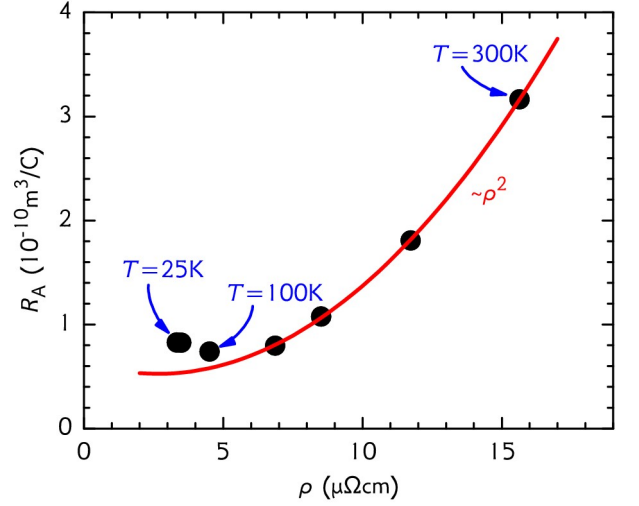


FIG. 6: Anomalous Hall coefficient R_A in dependence of the zero field resistivity ρ (black circles). Line: quadratic fit for temperatures above 100 K.

of this study. It is, however, worth to mention that Imort et al., in agreement with the conjectured skew scattering, reported $R_A \sim \rho$ for Co_2FeSi thin films, which naturally contain more defects than our high quality crystal [17]. Finally, it is worth to point out that the easily accessible and by temperature well distinct regimes of the AHE, suggests Co_2FeSi as an ideal test bed for theoretical treatments of the AHE.

In summary, we have investigated the electronic transport properties of Co_2FeSi in magnetic field. The temperature dependence of the resistivity is governed by electron-magnon scattering which exponentially freezes out over the substantial energy gap $\Delta \approx 100$ K at low temperatures and thus provides clear-cut evidence of Co_2FeSi being a HMF. This finding is corroborated by the magnetoresistance which is negative at high temperatures and positive at low temperatures. Furthermore, we observe that activation of ferromagnetic magnons at high temperatures $T \gtrsim \Delta$ has a strong impact on the anomalous Hall effect. In this high-temperature regime it is strongly temperature dependent and governed by Berry phase and side-jump deflection whereas at $T \lesssim \Delta$ it becomes temperature independent and is consistent with dominating skew scattering or intrinsic Lorentz force induced Hall effect.

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